Picosecond Structural Dynamics Probed by Time-resolved X-ray Diffraction

A.M. Lindenberg[1], I. Kang[1], S.L. Johnson[1], T. Missalla[5], P.A. Heimann[2], Z. Chang[3], M.M. Murnane[3], H.C. Kapteyn[3], P. H. Bucksbaum[3], J.S. Wark[4], R.W. Lee[5], R.W.Falcone[1]

[1] Department of Physics, 366 LeConte Hall, University of California, Berkeley, Berkeley, CA 94720, USA

[2]Advanced Light Source, Ernest Orlando Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

[3]Center for Ultrafast Optical Science, University of Michigan, Ann Arbor, MI 48109, USA

[4]Department of Physics, Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX13PU, United Kingdom

[5]Lawrence Livermore National Laboratory, PO Box 808, Livermore, CA 94551, USA

INTRODUCTION

Phase transitions and lattice dynamics in solids, structural changes in proteins, and chemical reactions occur on timescales of femtoseconds to picoseconds. Time-resolved x-ray diffraction offers a direct probe of these processes on the atomic scale. In this ongoing experiment, we investigate laser-induced structural dynamics in the semiconductor Indium Antimonide[1]. We report the direct observation of laser-induced coherent acoustic phonons, and probe in real time the transition from an ordered to a disordered state.

EXPERIMENT

The experiment was performed at beamline 7.3.3. The white beam is monochromatized to 5 keV by a Si (111) crystal and then diffracted off an asymmetrically-cut InSb (111) crystal onto the detector. The detector is a streak camera with 3 picosecond time resolution running in averaging mode[2]. A 1 kHz Ti:Sapphire laser system produces 150 femtosecond, 800 nm laser pulses, at 2.5 mJ/pulse. The laser is synchronized to the synchrotron RF with 2 ps jitter. One part of the laser pulse is split off to trigger a photoconductive switch which drives the sweep plates of the streak camera. A second part of the laser pulse is overlapped temporally and spatially with an x-ray pulse on the InSb crystal and initiates a structural change in the material. The crystal structure is then probed at later times by measuring the time-resolved diffracted intensity on the detector.

RESULTS

X-ray diffraction measurements were made at different rocking curve angles (the angular deviation from the Bragg condition) and at different laser fluences. In fig. 1, the normalized time-resolved diffraction efficiency is shown at an angle of +40 arcseconds, at a fluence of ~10 mJ/cm^2. It is observed that along with an initial drop in the diffracted intensity, the reflectivity oscillates with a period of ~15 picoseconds, indicative of fast, large-amplitude, coherent lattice oscillations.

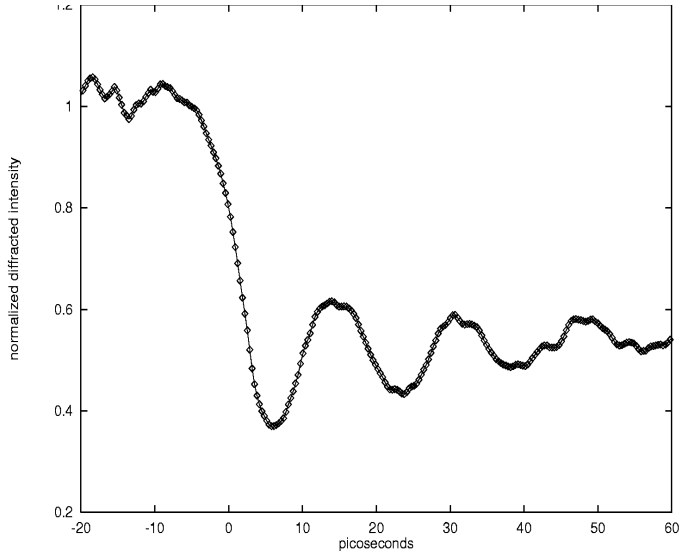

Figure 1. Normalized x-ray reflectivity on the large angle side of the rocking curve following ultrafast laser excitation.

We interpret this result in the following manner: On a timescale faster than the lattice can relax, an impulsively generated stress develops through either thermal heating of the lattice or direct acoustic deformation potential coupling of excited carriers to the lattice. This results in the excitation of coherent acoustic phonons across a range of wave vectors near the Brillouin zone center. Off the peak of the rocking curve, the wave vector of an acoustic phonon compensates for the mismatch between the reciprocal lattice vector and the scattering vector. This is similar to diffuse scattering experiments, except that the scattering is due to coherent rather than incoherent thermal vibrations, so that the intensity oscillates at the acoustic phonon frequency.

The acoustic phonon dispersion relation near the zone center is measured by recording the oscillation frequency for different angles relative to the Bragg peak (Fig. 2).

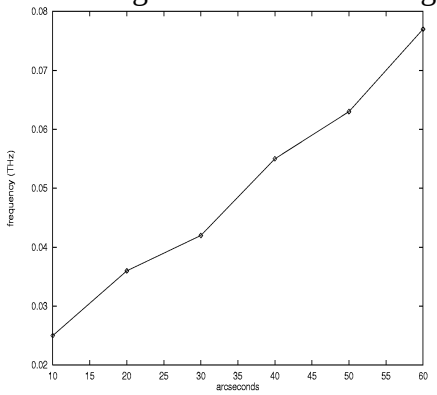

Figure 2. Oscillation frequency for different rocking curve angles, a measure of the phonon dispersion relation. The slope is proportional to the longitudinal acoustic sound velocity in the material.

From the slope of the above plot, we find the longitudinal acoustic sound velocity along the 111 direction to be 3 ± 1 km/sec in agreement with the known value of 3.7 km/sec.

The atomic displacements due to the coherent phonon oscillations can be estimated from the observed oscillation amplitude. We estimate an amplitude on the order of 10% of a lattice spacing, consistent with the incoherent acoustic phonon amplitude one would expect for InSb near its melting transition.

At higher laser fluences (>15 mJ/cm^2), near the damage threshold of the material, the diffracted intensity follows the lower fluence data for the first half period of an oscillation, but does not recover (Fig. 3). In effect, the atoms are impulsively excited out of the harmonic potential in which they normally reside, into a disordered state.

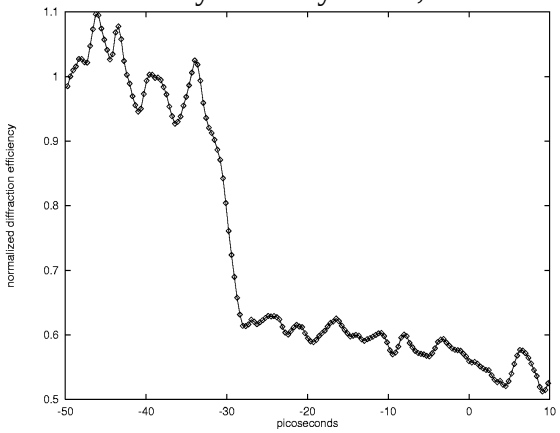

Figure 3. Normalized diffracted intensity on the larger angle side of the rocking curve, at a laser fluence ~ 15mJ/cm^2.

CONCLUSION

In this work, an ultrafast laser-induced phase transition in InSb was investigated using the method of time-resolved x-ray diffraction. We directly observe the excitation of large amplitude coherent lattice motion, and a transition to a disordered state. In addition, it is demonstrated that x-ray diffraction may be used in the time domain as a probe of phonon dynamics.

REFERENCES

[1]Larsson, J., Heimann, P.A., Lindenberg, A.M., Schuck, P.J., Bucksbaum, P.H., Lee, R.W., Padmore, H.A., Wark, J.S., Falcone, R.W. *Appl. Phys. A* **66**, 587 (1998).

[2]Larsson, J. Chang, Z., Judd, E., Schuck, P.J., Falcone, R.W., Heimann, P.A., Padmore, H.A., Kapteyn, H.C., Bucksbaum, P.H., Murnane, M.M., Lee, R.W., Machacek, A., Wark, J.S., Liu, X., Shan, B. *Optics Letters* **22**, 1012 (1997).

This work was supported by grants from the National Science Foundation and the Department of Energy through Lawrence Livermore National Laboratory and Lawrence Berkeley National Laboratory.

Principal investigator: R.W. Falcone, Department of Physics, University of California, Berkeley. Email: chairman@physics.berkeley.edu. Telephone: 510-642-8916.